# Penning Trap Measurements of the Masses of $^{133}$Cs, $^{87,85}$Rb, and $^{23}$Na with Uncertainties $\leq 0.2$ ppb

Michael P. Bradley, James V. Porto,* Simon Rainville, James K. Thompson, and David E. Pritchard
*Department of Physics and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*
(Received 19 April 1999)

We report new values from Penning trap single ion mass spectrometry for the atomic masses of $^{133}$Cs, $^{87}$Rb, $^{85}$Rb, and $^{23}$Na with uncertainties $\leq 0.2$ ppb, a factor of 100 improvement over the accuracy of previously measured values. We have found $M(^{133}\mathrm{Cs}) = 132.905\,451\,931(27)u$, $M(^{87}\mathrm{Rb}) = 86.909\,180\,520(15)u$, $M(^{85}\mathrm{Rb}) = 84.911\,789\,732(14)u$, and $M(^{23}\mathrm{Na}) = 22.989\,769\,280\,7(28)u$. These values are important for new ppb-level determinations of the molar Planck constant $N_A h$ and the fine-structure constant $\alpha$. With the measurement of $^{133}$Cs we have increased the mass range for sub-ppb measurements by a factor of 3. From $M(^{133}\mathrm{Cs})$ and other values we derive $\alpha^{-1} = 137.035\,992\,2(40)$.

PACS numbers: 32.10.Bi, 06.20.Jr, 07.75.+h

The fine-structure constant $\alpha$ is one of the most fundamental and best-measured quantities in physics, but the discrepancies between different precision measurements of $\alpha$ are noteworthy [1]. The most precise value $\alpha_{g-2}$ (uncertainty = 3.8 ppb ≡ 3.8 parts in $10^9$) has been obtained from the measured values of $(g - 2)$ for the electron and positron and complex QED calculations [2,3]. Unfortunately, the next most precise measurements of $\alpha$ (24 ppb using the quantum Hall effect [4,5] and 37 ppb via neutron interferometry [6]) disagree with the $(g - 2)$ value. Thus to test the unity of physics and QED there is a need for new ppb-level measurements to compare with $\alpha_{g-2}$.

A high-precision route to $\alpha$ founded on simple physics with no complex calculations is based on the relationship between $\alpha$ and the molar Planck constant $N_A h$ [7],

$$\alpha^2 = \frac{2R_\infty}{c}\frac{10^3}{M_p}\frac{m_p}{m_e}(N_A h)\,. \tag{1}$$

All the quantities linking $\alpha$ and $N_A h$ in Eq. (1) are accurately known: $R_\infty$ to 0.008 ppb [8,9], $m_p/m_e$ to 2 ppb [10], and $M_p$ (the proton mass in atomic units u) measured by our group to 0.5 ppb [11] (Van Dyck *et al.* have recently reported a value of $M_p$ accurate to 0.14 ppb [12]). Thus an independent measurement of $N_A h$ to 2 ppb would determine $\alpha$ to 1.4 ppb, a precision sufficient to test $\alpha_{g-2}$. The most complicated calculations for this method are the computations of the $2P_{1/2}$, $2P_{3/2}$, and $8D$ Lamb shifts to allow $R_\infty$ to be obtained from measurements of the $1S$-$2S$ and $2S$-$8D$ transitions in hydrogen [9]. The largest calculational uncertainty contribution to $R_\infty$ is 0.000 26 ppb due to the $8D$ Lamb shift.

$N_A h$ can be determined by measuring the velocity $v$ and de Broglie wavelength $\lambda_{dB} \equiv h/mv$ of a particle, since $\lambda_{dB} v = h/m = 10^3 N_A h/M$, where $m$ is the mass of the particle in SI units. Kruger *et al.* exploited this method using neutron interferometry to measure $h/m_n$ with 73 ppb precision [6]. Combining this with a precise measurement of $M_n$ (from Penning trap measurements of $M[^2\mathrm{H}]$, $M[^1\mathrm{H}]$ and $\gamma$-ray measurements of the nuclear binding energy of $^2$H [11,13,14]) gave a value of $\alpha$ with a precision of 37 ppb, again not sufficient to stringently test $\alpha_{g-2}$.

It appears that measurements of $N_A h$ of much higher precision can be made by combining accurate atom interferometry measurements of $h/m$ for alkalis with the ~0.1 ppb measurements of $M$ possible using Penning traps. At Stanford atom interferometry is used to measure $h/m_{Cs}$ via photon recoil [15] in terms of $\lambda_{D1}$ (recently measured to 0.12 ppb at Max-Planck-Institut Garching [16]). Although systematic effects in the photon recoil measurement currently limit the accuracy to 55 ppb, the high precision achieved (22 ppb in 4 h) [17] shows much promise. Moreover, advances in alkali Bose-Einstein condensation technology hold great promise for similar atom interferometry measurements on Na and Rb. With measurements of $h/m_{\mathrm{alkali}}$ in mind, we have measured the atomic masses of the stable alkali metal isotopes $^{133}$Cs, $^{87,85}$Rb, and $^{23}$Na. The previous mass accuracies [18] (see Table IV) would have limited the accuracy of $N_A h$ to several tens of ppb.

There are additional motivations for our measurements. New values of $N_A h$ at the few-ppb level in combination with measurements of $h$ (such as a recent 87 ppb measurement, expected to be improved by a factor of 10 [19]) can yield values of $N_A$ with ppb-level accuracy. Since $N_A$ links the atomic and SI units of mass this is of great interest [7]. Furthermore, Cs and Rb are used as reference masses for measurements of heavy radioactive nuclei which are important for modeling astrophysical heavy element formation [20,21].

We obtain absolute atomic masses $M$ from mass ratios relating the unknown mass to the atomic mass standard $^{12}$C. Ion mass ratios are obtained from ratios of measured cyclotron frequencies $\omega_c = qB/mc$, where $m$ and $q$ are, respectively, the ion mass and charge. We make precise cyclotron frequency measurements on a single ion confined in an orthogonally compensated Penning trap [22] of characteristic size $d = 0.55$ cm placed in the $B = 8.5$ T field of a superconducting magnet. Use of a trapped


0031-9007/99/83(22)/4510(4)$15.00

single ion takes advantage of long observation times in the absence of perturbations from ion-ion interactions and allows sub-ppb precision.

An ion in our Penning trap has three normal modes of motion: trap cyclotron, axial, and magnetron, with frequencies $\omega_c' \gg \omega_z \approx 2\pi \times 160$ kHz $\gg \omega_m$, respectively. The free-space cyclotron frequency $\omega_c$ is obtained from the following expression (invariant with respect to trap tilts and ellipticity) [22]:

$$\omega_c = qB/mc = \sqrt{(\omega_c')^2 + (\omega_z)^2 + (\omega_m)^2}. \qquad (2)$$

We detect axial motion by coupling to a dc SQUID via a $Q = 5 \times 10^4$ superconducting resonant transformer. Detection damps the axial motion at a rate $\gamma_z \sim 1\ \mathrm{s}^{-1}$ and cools it to 4 K. The amplitude and phase of the undamped trap cyclotron mode are measured via coupling to the axial mode established by an rf quadrupole field at frequency $\omega_\pi = \omega_c' - \omega_z$ [23]. (This coupling is also used to cool the radial modes.) We obtain $\omega_c'$ from the phase accumulated in the cyclotron mode in a given time. Using the measured values of $\omega_c'$ and $\omega_z$ we calculate $\omega_m \approx (\omega_z^2/2\omega_c')(1 + 9/4 \sin^2\theta_m)$, where $\theta_m = 0.16°$ is the measured angle between the $B$ field and trap axes (the effect of $\theta_m$ on a mass ratio is at most 0.002 ppb).

We measured the free-space cyclotron frequency ratios $r \equiv \omega_{c2}/\omega_{c1}$ listed in Table I. A cyclotron frequency ratio $r$ was determined by a run of alternately measured clusters of $\omega_c$ of each of the two ions during the period from 1:30–5:30 A.M. when the nearby electrically powered subway was not running (Fig. 1). The measured free-space cyclotron frequencies exhibited a common slow drift. We fit a common polynomial $\Omega(t)$ plus a frequency difference to the data. From this we obtained the frequency ratio $r_n$ and the uncertainty $\sigma_n$ for a single night. The average order of $\Omega(t)$ was 3 and was chosen using the F-test criterion [24] as a guide.

Excluding the $Cs^{++}/C_5H_6^+$ data, the distribution of residuals from the polynomial fits had a Gaussian center with a standard deviation $\sigma_{\mathrm{resid}} = 0.28$ ppb and a background ($\approx 2\%$ of the points) of non-Gaussian outliers, as in our earlier measurements [11]. As in [11] we chose to handle the non-Gaussian outliers using a robust statistical method to smoothly deweight them [25]. The observed random fluctuations of the measured free-space cyclotron frequency should limit the precision of a one-night (4 h) measurement of $r_n$ to $\sim 0.1$ ppb. These fluctuations are due primarily to variations of the magnetic field; however, for heavy ions with low $\omega_c$ uncertainty in $\omega_z$ can contribute significantly to uncertainty in $\omega_c$. Thus stability of the trapping voltage is important. Our trap voltage exhibits long-term rms fluctuations of only 100 ppb and so contributes below 0.1 ppb uncertainty to a night's measurement. Observed variations in the axial frequency $\sim 20$ mHz are dominated by measurement error and contribute at most $\sim 0.25$ ppb to a single cluster for $Cs^{++}/C_5H_6^+$ (the residuals for $Cs^{++}/C_5H_6^+$ had $\sigma_{\mathrm{resid}} = 0.44$ ppb mainly because of this).

TABLE I. Measured ion cyclotron frequency ratios, corrected for systematics.

| $A/B$ | $\frac{\overline{\omega}_c}{2\pi}$ (MHz) | Nights | $\omega_c[A]/\omega_c[B]$ |
|---|---|---|---|
| $^{133}Cs^{+++}/CO_2^+$ | 2.968 | 5 | 0.992 957 580 983(135) |
| $^{133}Cs^{++}/C_5H_6^+$ | 1.977 | 4 | 0.993 893 716 487(427) |
| $^{87}Rb^{++}/C_3H_8^+$ | 2.994 | 2 | 1.013 992 022 591(266) |
| $^{87}Rb^{++}/C_3H_7^+$ | 3.028 | 3 | 0.990 799 127 824(174) |
| $^{85}Rb^{++}/C_3H_7^+$ | 3.064 | 2 | 1.014 106 122 230(164) |
| $^{85}Rb^{++}/C_3H_6^+$ | 3.100 | 2 | 0.990 367 650 976(285) |
| $^{23}Na^+/C_2^+$ | 5.578 | 2 | 1.043 943 669 690(076) |
| $^{23}Na^{++}/C^+$ | 11.155 | 2 | 1.043 944 716 614(098) |

As shown in Table I we measured each frequency ratio on more than a single night. For ratios involving Cs and Rb the measured $r_n$ were distributed with scatter $> \sigma_n$ ($\chi_\nu^2 \approx 5$). By contrast $\chi_\nu^2 \approx 0.8$ for ratios involving Na. All earlier data taken using this apparatus [11] did not exhibit these excess night-to-night variations. Possible sources of this excess scatter are presented in Table II and discussed below.

The axial frequency $\omega_z$ is "pulled" by its coupling to the detector by an amount $\Delta\omega_z = (\gamma_z/\gamma_0)(\omega_z - \omega_0)$, where $\omega_0$ and $\gamma_0/2\pi = 3$ Hz are the detector resonance frequency and FWHM. We adjusted $\omega_z$ to be equal to the slowly drifting $\omega_0$ ($\sim 50$ mHz/h), measured before each alkali ion cluster. We then used the measured values of $\omega_z$ and $\omega_0$ ($|\omega_z - \omega_0|$ typically $< 100$ mHz) to correct the measured $\omega_z$ for the remaining pulling shift. Table II gives the rms difference in $r_n$ for each night computed before and after applying this correction. The $\sim 30\%$ accuracy of our corrections implies that frequency pulling represents an error in our final ratios below 0.03 ppb and was not the source of the excess night-to-night fluctuations of $r_n$.

Relativistic mass variation and spatial imperfections $\Delta\vec{B} = -(B_2/2)\rho^2\hat{z}$ and $\Delta V/V_{\mathrm{trap}} = -(3/16)(C_4/d^4)\rho^4$ [22] of the trap fields lead to radius-dependent shifts of the cyclotron frequency

$$\frac{\Delta\omega_c}{\omega_c} = \left(-\frac{\omega_c'^2}{2c^2} - \frac{B_2}{2B_0} + \frac{3\omega_m C_4}{2\omega_c d^2}\right)\rho^2. \qquad (3)$$

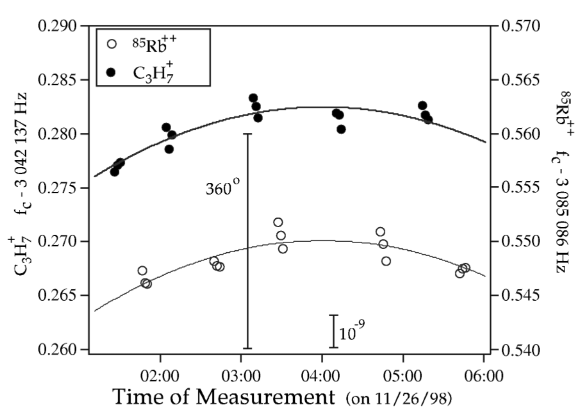


FIG. 1. Typical night of data. The solid line is a second order polynomial fit to the data. The 360° bar shows the magnitude in Hz of a 360° error in phase unwrapping.

TABLE II. Random uncertainties ($\sigma$) and systematic shifts ($\Delta$) for frequency ratios $r$ in ppt (parts per $10^{12}$).

| | $\frac{Cs^{+++}}{CO_2{}^+}$ | $\frac{Cs^{++}}{C_5H_6{}^+}$ | $\frac{Rb^{++}}{C_3H_x{}^+}$ | $\frac{Na^+}{C_2{}^+}$ | $\frac{Na^{++}}{C^+}$ |
|---|---|---|---|---|---|
| $\sigma(r_n)/r$ axial frequency pulling | 128 | 64 | 62 | 11 | 29 |
| $\Delta r/r$ relativity[a] | 2 | 1 | 7 | 4 | 79 |
| $\Delta r/r$ $B_2$[a] | −2 | −4 | 9 | −13 | −38 |
| $\Delta r/r$ $C_4$[a] | −1 | −4 | 2 | −9 | −6 |
| $\Delta r/r$ different positions[b] | 2 | 1 | 3 | −19 | −38 |

[a]These shifts in the ratio are due to systematic differences in the cyclotron radii ($\Delta\rho/\rho \approx 1\%$ to 7% with $\langle\rho\rangle = 230$ $\mu$m). $B_2/B_0 = -1 \times 10^{-6}/\text{cm}^2$ and $|C_4| < 10^{-4}$.
[b]Shifts due to differential axial displacement (the largest being $\Delta z \approx 0.24$ $\mu$m for $Na^{++}/C^+$) combined with the measured $B_1/B_0 = -1.6 \times 10^{-6}$ cm$^{-1}$.

For our trap $B_2/B_0 = 10^{-6}$ cm$^{-2}$ and $|C_4| < 10^{-4}$. Systematic differences in cyclotron orbit radii gave negligible systematic shifts in $r$ except for $Na^{++}/C^+$ ($\Delta r/r =$ 0.035 ppb). We adjusted the final ratio for this shift. To reflect uncertainty in the absolute radius $\rho$ we assigned the adjustment a 50% uncertainty added in quadrature with the other uncertainties.

Table II gives the systematic shifts in $r$ for differential displacements of ions from the geometric center of the trap (due to the electric field from fixed charge patches) in combination with $B$-field inhomogeneities. The $z$ component of the patch electric field was measured with <10% uncertainty by applying offset potentials and measuring the quadratic shifts in $\omega_z$, as described in [26]. $Na^{++}/C^+$ experienced the largest systematic shift $\Delta r/r = 0.08$ ppb. We adjusted all ratios for this systematic shift and assigned the adjustment a 100% uncertainty added in quadrature with the statistical uncertainty to reflect uncertainty concerning the radial component of the charge patch electric field.

The systematic shifts in Table II are constant from night to night and also are much too small to explain the observed night-to-night scatter.

To test the hypothesis that the excess night-to-night fluctuations were due to improper choice of fit order for $\Omega(t)$ we computed $r_n$ by a completely different method. A value of $r$ was calculated for each cluster in a night by taking the ratio of its frequency to the frequency obtained by linear interpolation between the two neighboring clusters of the other ion type. The values of $r$ for each cluster were averaged to yield a single value of $r_n$ for each night. This "piecewise linear" method and the polynomial fits gave similar night-to-night variations in $r_n$, suggesting that the variations were not polynomial fitting artifacts.

We calculated the weighted average, for the polynomial fit and piecewise linear methods separately, of the values of $r_n$. There was no statistically significant difference between these averages $\bar{r}_{\text{poly}}$ and $\bar{r}_{\text{pl}}$ except for $Cs^{++}/C_5H_6{}^+$ (0.57 ppb disagreement) due to one night with anomalously constant magnetic field, which gave a small uncertainty and hence significantly higher weight to that night's $r_n$ computed with the piecewise linear method. To account for any such discrepancies we quote the average $\bar{r} = (1/2)(\bar{r}_{\text{poly}} + \bar{r}_{\text{pl}})$ and a final uncertainty $\bar{\sigma}^2 = \tilde{\sigma}^2_{\text{poly}} + [(\bar{r}_{\text{poly}} - \bar{r}_{\text{pl}})/2]^2$, where $\tilde{\sigma}_{\text{poly}}$ is the uncertainty in $\bar{r}_{\text{poly}}$ after the rescaling described below.

Ultimately we did not discover the origin of the night-to-night fluctuations of $r_n$ for Cs and Rb. We therefore increased the quoted statistical uncertainties to ensure that they reflect the observed night-to-night scatter. For $Cs^{++}/C_5H_6{}^+$ and $Cs^{+++}/CO_2{}^+$, we increased each night's statistical uncertainty until $\chi^2_\nu = 1$ (from 6.6 and 4.9, respectively) for each ratio separately. For the $^{87,85}$Rb ratios we assumed that the night-to-night fluctuations were drawn from a common statistical distribution since the Rb measurements all had similar $m/q$. Therefore, we increased the statistical uncertainty on all the Rb $r_n$ by a common factor to reduce the overall Rb $\chi^2_\nu$ from 4.7 to 1. The resulting increased statistical uncertainties for $\bar{r}_{\text{poly}}$ are the $\tilde{\sigma}_{\text{poly}}$ used above. The Na ratios had no significant night-to-night fluctuations and required no adjustment of their uncertainties.

TABLE III. Measured alkali masses from different routes.

| Species | Ref. ion | Mass (u) | $\sigma_M/M$ (ppb) | $\chi^2_\nu$ |
|---|---|---|---|---|
| $^{133}$Cs | $CO_2{}^+$ | 132.905 451 931(22) | 0.17 | 0.003 |
| | $C_5H_6{}^+$ | 132.905 451 934(57) | 0.43 | |
| $^{87}$Rb | $C_3H_8{}^+$ | 86.909 180 540(24) | 0.28 | 0.92 |
| | $C_3H_7{}^+$ | 86.909 180 511(17) | 0.19 | |
| $^{85}$Rb | $C_3H_7{}^+$ | 84.911 789 737(15) | 0.18 | 0.45 |
| | $C_3H_6{}^+$ | 84.911 789 717(25) | 0.30 | |
| $^{23}$Na | $C_2{}^+$ | 22.989 769 278 9(17) | 0.07 | 2.97 |
| | $C^+$ | 22.989 769 283 7(22) | 0.09 | |

TABLE IV. Measured neutral alkali masses.

| Species | MIT Mass (u) | 1993 Mass (u) [18] |
|---|---|---|
| $^{133}$Cs | 132.905 451 931(27) | 132.905 447 000(3000) |
| $^{87}$Rb | 86.909 180 520(15) | 86.909 185 800(2800) |
| $^{85}$Rb | 84.911 789 732(14) | 84.911 792 400(2700) |
| $^{23}$Na | 22.989 769 280 7(28) | 22.989 769 660 0(2600) |

After correcting for a 93(1) $\mu$Hz$/e$ image-charge shift, we convert ion frequency ratios $r$ to neutral mass ratios by accounting for missing electrons [18] and ionization and chemical binding energies [27]. These adjustments contribute $\leq$0.03 ppb uncertainty to the final neutral mass ratios. We neglect vibrational/rotational excitation due to ionization. Vibrational energy decays in msec (except for homonuclear species like $C_2{}^+$, where the decay time $\sim$1 min). Rotational energies ($\sim$meV) can be ignored.

From the neutral mass ratios we obtained a set of neutral mass difference equations. We added to this the mass difference equations used to determine the atomic masses in [11]. Solution of this overdetermined set of linear equations gave the neutral masses of the alkali metals (see Table IV) with uncertainties $\sigma_{\rm od}$ as well as the previously published neutral masses with $\chi^2_\nu = 0.83$. The previously published masses were essentially unchanged and so are not reported. Uncertainties in $M[^{16}\mathrm{O}]$ and $M[\mathrm{H}]$ contributed $<$0.1 ppb uncertainty to the alkali masses.

The use of two distinct reference ions gave a check on systematics by providing two independent values for each neutral mass (Table III). For Rb and Cs $\chi^2_\nu$ is less than 1. However, because of the larger uncertainty on $M[\mathrm{Cs}]$ from $\mathrm{Cs}^{++}/\mathrm{C_5H_6}^+$ we quote a final uncertainty of 0.20 ppb $> [\sigma_{\rm od}(\mathrm{Cs}) = 0.16$ ppb]. For $^{87,85}$Rb we quote $\sigma_{\rm od}(^{87,85}\mathrm{Rb})$ as the final uncertainties. For the neutral masses from $\mathrm{Na}^{++}/\mathrm{C}^+$ and $\mathrm{Na}^+/\mathrm{C_2}^+$, the statistical uncertainties are 0.09 and 0.07 ppb, respectively. The 0.2 ppb disagreement of the two values may be evidence for a systematic at the 0.1 ppb level. To reflect this we assigned $M[^{23}\mathrm{Na}]$ a 0.12 ppb uncertainty $>[\sigma_{\rm od}(\mathrm{Na}) =$ 0.06 ppb] which spans both independent measurements.

Table IV gives final values for $M[^{133}\mathrm{Cs}]$, $M[^{87}\mathrm{Rb}]$, $M[^{85}\mathrm{Rb}]$, and $M[^{23}\mathrm{Na}]$ obtained from the solution of the mass difference equations with uncertainties from the above discussion. Also included in Table IV are alkali masses from the 1993 mass evaluation [18]. Our values differ from the 1993 values by $\sim 1.5\sigma_{1993}$. Our value for $M(^{133}\mathrm{Cs})$ lies within the uncertainty of the recent measurement of $M(^{133}\mathrm{Cs})$ reported by the SMILETRAP Collaboration [28]. Using $R_\infty$ [8], $m_p/m_e$ [10], the preliminary value of the photon recoil shift [17], $f_{D1}$ for the photon recoil transition [29], and our values for $M(^{133}\mathrm{Cs})$ and $M_p$ we obtain $\alpha^{-1} = 137.035\,992\,2(40)$.

We are grateful to Roland Nguyen for winding our $Q \approx 5 \times 10^4$ detector coil. This work was supported by the National Science Foundation (Grant No. PHY-9514795), a NIST Precision Measurements Grant (Grant No. 60NANB8D0063), and the Joint Services Electronics Program (Grant No. DAAH04-95-1-0038).

*Currently at NIST, Gaithersburg, MD 20899-8421.

[1] T. Kinoshita, Rep. Prog. Phys. **59**, 1459 (1996).
[2] T. Kinoshita, IEEE Trans. Instrum. Meas. **46**, 108 (1997); Phys. Rev. Lett. **75**, 4728 (1995).
[3] R. S. Van Dyck, Jr., P. B. Schwinberg, and H. G. Dehmelt, Phys. Rev. Lett. **59**, 26 (1987).
[4] M. E. Cage *et al.*, IEEE Trans. Instrum. Meas. **38**, 284 (1989).
[5] A. Jeffery *et al.*, IEEE Trans. Instrum. Meas. **46**, 264 (1997).
[6] E. Kruger, W. Nistler, and W. Weirauch, Metrologia **35**, 203 (1998).
[7] B. N. Taylor, Metrologia **31**, 181 (1994).
[8] C. Schwob *et al.*, Phys. Rev. Lett. **82**, 4960 (1999).
[9] Th. Udem *et al.*, Phys. Rev. Lett. **79**, 2646 (1997).
[10] D. L. Farnham, R. S. Van Dyck, Jr., and P. B. Schwinberg, Phys. Rev. Lett. **75**, 3598 (1995).
[11] F. DiFilippo, V. Natarajan, K. R. Boyce, and D. E. Pritchard, Phys. Rev. Lett. **73**, 1481 (1994).
[12] R. S. Van Dyck, Jr., D. L. Farnham, S. Zafonte, and P. B. Schwinberg, *ICAP XVI Abstracts, Windsor, 1998* (University of Windsor, Windsor, 1998), pp. 294–295.
[13] R. S. Van Dyck, Jr., D. L. Farnham, and P. B. Schwinberg, *Abstracts of the American Physical Society, 1993* (American Physical Society, Ridge, NY, 1993).
[14] G. L. Greene, E. G. Kessler, Jr., R. D. Deslattes, and H. Börner, Phys. Rev. Lett. **56**, 819 (1986).
[15] D. Weiss, B. Young, and S. Chu, Phys. Rev. Lett. **70**, 2706 (1993); Appl. Phys. B **59**, 217 (1994).
[16] Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Phys. Rev. Lett. **82**, 3568 (1999).
[17] B. C. Young, Ph.D. thesis, Stanford, 1997.
[18] G. Audi and A. G. Wapstra, Nucl. Phys. **A565**, 1 (1993).
[19] E. R. Williams, R. L. Steiner, D. B. Newell, and P. T. Olsen, Phys. Rev. Lett. **81**, 2404 (1998).
[20] H. Stolzenberg *et al.*, Phys. Rev. Lett. **65**, 3104 (1990).
[21] B. Fogelberg *et al.*, Phys. Rev. Lett. **82**, 1823 (1999).
[22] L. S. Brown and G. Gabrielse, Rev. Mod. Phys. **58**, 233 (1986).
[23] E. A. Cornell, R. M. Weisskoff, K. R. Boyce, and D. E. Pritchard, Phys. Rev. A **41**, 312 (1990).
[24] P. R. Bevington and D. K. Robinson, *Data Reduction and Error Analysis for the Physical Sciences* (McGraw-Hill, Boston, 1992), 2nd ed.
[25] P. J. Huber, *Robust Statistics* (Wiley, New York, 1981).
[26] V. Natarajan, K. R. Boyce, F. DiFilippo, and D. E. Pritchard, Phys. Rev. Lett. **71**, 1998 (1993).
[27] S. G. Lias *et al.*, in *NIST Chemistry WebBook*, NIST Standard Reference Database No. 69, edited by W. G. Mallard and P. J. Linstrom (NIST, Gaithersburg, MD, 1998) (http://webbook.nist.gov).
[28] C. Carlberg, T. Fritioff, and I. Bergstrom, preceding Letter, Phys. Rev. Lett. **83**, 4506 (1999).
[29] Thomas Udem (private communication).